

# Dynamics of Aqueous Foam Drops

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We develop a model for the nonlinear oscillations of spherical drops composed of aqueous foam. Beginning with a simple mixture law, and utilizing a mass-conserving bubble-in-cell scheme, we obtain a Rayleigh-Plesset-like equation for the dynamics of bubbles in a foam mixture. The dispersion relation for sound waves in a bubbly liquid is then coupled with a normal modes expansion to derive expressions for the frequencies of eigenmodal oscillations. These eigenmodal (breathing plus higher-order shape modes) frequencies are elicited as a function of the void fraction of the foam. A Mathieu-like equation is obtained for the dynamics of the higher-order shape modes and their parametric coupling to the breathing mode. The proposed model is used to explain recently obtained experimental data.

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## I. INTRODUCTION

Foams and froths are ubiquitous in nature and industry. They are the signature of vigorous, gas-entraining mixing processes in liquids. A minimalist conception of a foam would consist of a gas confined as bubbles within a liquid host. The aqueous foams considered here are composed of surfactant-bearing water and air bubbles. A comprehensive review of foam theories and applications can be found in the textbooks by Edwards *et al* [1], Exerowa and Kruglyakov [2], and the article by Kraynik [3].

Theoretical treatments of the unique rheology of foams go back at least to Mallock [4], who was motivated to explain the common observation that “A tumbler containing a frothy liquid gives a dull sound when struck”. Mallock showed that the sound speed for intermediate void fractions was actually lower than its value for either the wet limit,  $c_{water} = 1500$  m/s, or the dry limit,  $c_{air} = 340$  m/s. This result has been borne out by a century of subsequent work on bubbly liquids [5]–[9], and, as seen later, leads to key insights into the free vibrations of foam drops.

The present work is motivated by the continuing need to measure, understand, model and eventually predict foam mechanics and rheology for wet or dry foams. To this end, two of the present authors recently described a non-contact technique in which small samples of foam (“foam drops”, see Fig. 1) were acoustically levitated and excited into resonance by modulating the levitation field [10]. Our technique utilizes acoustic levitation to provide both non-contact positioning and static and oscillatory excitation of foam drops. By measuring the quadrupole eigenfrequency of a 3.8 mm radius foam drop to be 63 Hz, we inferred a shear modulus of roughly 73–78 Pa for a relatively dry foam. This value compared favorably with experimentally determined moduli utilizing more traditional contact-based techniques ([11]–[16]). The same technique has proven successful for determination of the surface rheological properties of single-phase liquid drops [17]–[19].

The primary advantages of this acoustic levitation technique are its elimination of the requirement for sample contact containment, its ability to test foams of arbitrary gas volume fraction, and its ability to excite both shear and dilatational motion. The technique relies on a suitable physical model for the dynamic response of spheroidal foam drops. The heart of this model is the theoretical description of the foam material. Our earlier work [10] modelled the foam as an effective solid clas-

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tic medium. While successful in describing the small-amplitude oscillations of a dry foam with a fixed lattice of bubbles, such a material description has certain disadvantages. First, a foam is only solid-like for high gas volume fractions and small amplitude motion. Second, such an effective elastic medium theory only implicitly incorporates the effect of varying gas volume fraction via the effective density of the medium, and it cannot capture the physics of a bubbly mixture. Finally, the gas volume fraction is a dynamic quantity, and during dilatational motion of significant amplitude it cannot be treated as a material constant. We thus need a model capable of describing wet foams.

The subject of the present paper is a theoretical investigation of the dynamics of wet foam drops. By using a bubbly-fluid approach we obtain a model for the time-dependent response of a foam drop which retains the nonlinearity of the bubbles response. By linearizing the equations we elucidate the resonance frequencies for the breathing and higher-order eigenmodes of a drop. We treat the coupling of shape and breathing modes, and investigate numerically the nonlinear equations obtained for the time evolution and interaction of the modal oscillations. A key feature is the inclusion of a time-varying void fraction in the equations.

We begin in the next section with an argument for the relevant physics that must be included in the model.

## II. FOAM MECHANICS, RHEOLOGY, AND DROP DYNAMICS

One of the most important characteristic parameters of a foam is its gas volume fraction  $\alpha_g$ , or more commonly the “void fraction.” A foam’s thermodynamic, mechanical, acoustical and rheological properties are sensitive functions of the void fraction. Three regimes of foam morphology are typically identified. A “wet foam” (approximately  $0 < \alpha_g < 0.5$ ) is essentially a bubbly liquid. The individual bubbles are free to move about within the liquid. Wet foams cannot support shearing motion, except at the surface of the individual bubbles. A “transitional” or “critical foam” (approximately  $0.5 < \alpha_g < 0.7$ ) is comprised of bubbles whose dynamics are strongly interacting, and whose surfaces may be in mechanical contact with each other. This regime may be usefully thought of as a phase transition between a liquid and solid-like state. A critical void fraction marks the point at which a foam begins to possess solid-like properties, such as shear wave propagation and yield stress. The critical void fraction for three-dimensional foams is approximately 0.67, which geometrically corresponds to random close packing of bubbles. Finally, a “dry foam” is the commonly encountered state in which the bubbles, at least for low to moderate straining rates, have a fixed position in a lattice. Such foams behave as viscoelastic solids for sufficiently small straining rates. However, a

dry foam may flow as a liquid when strained beyond a critical point.

Theoretical investigations of rheological properties begin with Derjaguin, [20] who derived an expression for the shear modulus of an idealized dry foam and showed that it was linearly proportional to the foam capillary pressure. This result implies that a foam’s modulus should scale as  $\alpha_g^{-1/3}$ . Subsequent theoretical and numerical work has concentrated primarily on two-dimensional geometric models limited to dry foams. The reader is referred to references [21]–[26] for examples of the groundbreaking work in this area and to reference [27] for a comprehensive review. Several theoretical models have addressed the unique rheological dependence on void fraction of foams. Bolton and Weaire [28] introduced a two-dimensional model that predicts the vanishing of the shear modulus at a critical void fraction. This model is strictly valid in the dry limit. In contrast, a static but bubble-based two-dimensional molecular dynamics simulation [29] captures the transition features using wet-limit assumptions. The model assumes spherical bubbles that resist deformation because of their Laplace pressure. Among other things, the model predicts the vanishing of the shear modulus, but the scaling behavior near the transition is different than that found for dry two-dimensional models.

It is interesting to qualitatively consider the dynamics of foam drops in the limiting cases of wet, critical and dry. We consider a foam drop surrounded by a gas to simplify the situation. To engage in even a brief discussion, we must draw a distinction between the breathing or monopole mode and the higher-order shape or multipole modes, since such motions are qualitatively different as well. First we consider breathing mode oscillations. The restoring force for perturbations from the drop’s equilibrium volume is provided by the internal pressure of the individual bubbles within the drop, which will expand and contract when the drop volume is externally forced. Surface tension plays a small role, since the Laplace pressure  $2\sigma/R_0$  for a drop of radius  $R_0$  is much smaller than the ambient or atmospheric pressure. The mass is that of the liquid between the bubbles. Dissipation is provided by the bulk fluid motion, the surface fluid motions at the drop surface and at the individual bubble surfaces, and also by heat transfer and acoustic radiation of the individual oscillating bubbles [30]. For critical and dry foam drops, a primary difference is that surface tension becomes more important as a restoring force because of the many thin film fluid connections which form inside the drop. The mass continues to decrease as the void fraction increases. It is difficult to make any general statement about the effect on dissipation due to increasing void fraction, except to say that dissipative effects are growing relative to inertial effects.

For shape oscillations of wet foam drops, the restoring force for perturbations from the drop’s equilibrium shape is surface tension acting at the drop interface.

Since surface-active agents are present, a local Marangoni restoring force due to gradients in surface tension also contributes. The effective mass is once again the mass of the liquid component. The dissipation is more strongly affected by the surface terms at the drop interface than for the monopole case, and the thermal and acoustic bubble dissipation terms are negligible. For shape oscillations of critical and dry foam drops, the internal thin film fluid connections add stiffness to the drop, plus allowing the possibility of torsional multipole modes. As for the monopole case, the mass is decreasing, and dissipation is again ambiguous as the void fraction increases.

The discussion above has implicitly assumed that the internal pressure of the mixture inside the drop is uniform (except for the Laplace pressure contribution to the (again assumed uniform) interior bubble pressure). This will hold true until either the wavelength of incident sound is short compared to the drop radius  $R$  (which will never happen during standing wave acoustic levitation), or the velocity of the drop interface  $\dot{R}$  approaches the speed of sound in the mixture, which is also unlikely. Uniform mixture pressure implies that all bubbles oscillate essentially in phase unless there are wide disparities in the bubble size distribution, or nonlinear effects dominate.

For both breathing and shape modes, foam drops near the critical void fraction will experience a sort of mode dispersion as bubbles begin to strongly interact with their nearest neighbors. Energy initially concentrated in a single global drop mode will be dispersed into motion of small collections of interacting bubbles. The result may well be an "apparent" increase in damping of the global observable drop modal oscillation. While this is highly conjectural, we believe we have indeed observed the apparent damping effect for forced quadrupole oscillations of near-critical foam drops. For the same liquid constituent, the quadrupole mode for both wet and dry foam drops was underdamped and thus a resonance was observable. However, for near critical void fractions the resonance was unobservable. A detailed investigation of this effect is a topic for future work.

Thus we turn our attention to a more dynamic and bubble-based description of a foam, and in so doing we explicitly incorporate the fact that we utilize acoustics and acoustic levitation of bounded foam drops in our experiments. We wish to improve upon such existing models by considering 3-dimensional cases, and by explicitly incorporating bubbly fluid dynamics and acoustic wave propagation in our model. We begin here by introducing a three-dimensional (spherically- or axisymmetric) model for the eigenmodal oscillations of spheroidal foam drops in the wet limit.

### III. WAVE EQUATION FOR WET FOAM

In this section we follow [8] to introduce the wave equation for an aqueous foam in the wet limit. For the envisioned applications of this analysis, the liquid may be treated as incompressible. Viscous dissipation as it appears in the normal stress balance for individual bubble oscillations is included. Thermal and acoustic dissipation are neglected. Likewise, we do not consider processes such as bubble coalescence, breakup, or dissolution which affect the number of bubbles and/or their equilibrium size.

To avoid confusion with the standard notation for velocity potentials (note that several authors use  $\phi$  to denote the void fraction), let  $\alpha_l$  and  $\alpha_g$  be the fractional volume concentration, and  $\rho_l$  and  $\rho_g$  the density of the liquid and the gas, respectively. Then the density  $\rho$  of the two-phase mixture is given by

$$\rho = \alpha_l \rho_l + \alpha_g \rho_g, \quad (1)$$

where  $\alpha_l + \alpha_g = 1$ . With  $\rho_g \ll \rho_l$  and the assumption that all bubbles have the same radius  $a$ , we obtain the following approximation for the mixture density:

$$\rho \approx \rho_l(1 - \alpha_g), \quad \alpha_g = \frac{4}{3}\pi a^3 n, \quad (2)$$

where  $n$  is the number of bubbles per unit volume of the mixture.

The dynamics of the bubbles in a foam may be treated with a spherically-symmetric bubble-in-cell scheme. Accordingly, each single bubble in the bubbly liquid is placed in the center of a spherical cell with the radius  $A$  chosen in such a way as to cover the whole bubbly mixture by cells.

$$A = \frac{a}{\alpha_g^{1/3}}. \quad (3)$$

It should be noted that  $A, a, \alpha_g$  are variable in time and space, and  $a < A \ll R$  typically. Here,  $R$  is the radius of the drop.

Bernoulli's integral for spherically-symmetric incompressible liquid motion in the cell may be written in the following form:

$$p = p_a - \rho_l \left( a\ddot{a} + \frac{3}{2}\dot{a}^2 \right) + \frac{\rho_l}{r} (a^2\dot{a})_t - \frac{\rho_l a^4 \dot{a}^2}{2r^4}, \quad (4)$$

where dots denotes time derivatives,  $r$  is the radial coordinate with the origin at the bubble center,  $\nu$  is the shear kinematic viscosity of the liquid,  $p_a$  is the liquid pressure at the bubble wall, which is related to the gas bubble pressure  $p_g$  by the formula

$$p_g = p_a + \frac{2\sigma}{a} + \frac{4\rho_l \nu \dot{a}}{a}. \quad (5)$$

Here,  $\sigma$  is the surface tension coefficient and  $\nu$  is the kinematic viscosity of the liquid. Often, it is treated as an "effective" viscosity which accounts for dissipation [8].

Taking into account that the liquid pressure at the cell boundary is equal to the pressure of the two-phase mixture  $p$  [5] one can obtain the following generalized Rayleigh-Plesset equation for radial bubble motion in the cell:

$$\left(1 - \alpha_g^{1/3}\right) a \ddot{a} + \frac{3}{2} \left(1 - \frac{4}{3} \alpha_g^{1/3} + \frac{1}{3} \alpha_g^{4/3}\right) \dot{a}^2 = \frac{p_a - p}{\rho_l}. \quad (6)$$

The gas pressure in the bubble is assumed to be uniform and variable in time and may be calculated via a polytropic approximation.

$$p_g = \left(p_0 + \frac{2\sigma}{a_0}\right) \left(\frac{a}{a_0}\right)^{-3\kappa}, \quad (7)$$

where  $\kappa$  is the polytropic exponent ( $\kappa = \gamma_g$  for adiabatic and  $\kappa = 1$  for isothermal oscillations of the bubbles, where  $\gamma_g$  denotes the gas adiabatic exponent).

Next we outline the approach to obtaining the mixture wave equation. The conservation equations for the number of bubbles, the mass of the mixture, and momentum of the mixture are first linearized about the unperturbed state of the mixture. For this purpose we introduce small perturbations  $p'$ ,  $\rho'$ ,  $\vec{v}'$ ,  $n'$ , and  $a'$  of the equilibrium values  $p_0$ ,  $\rho_0 = \rho_l(1 - \alpha_{g0})$ ,  $\vec{v}_0 = 0$ ,  $n_0$ , and  $a_0$  such that  $p = p_0 + p'$ ,  $\rho = \rho_0 + \rho'$ ,  $\vec{v} = \vec{v}'$ ,  $n = n_0 + n'$ , and  $a = a_0 + a'$ . Then we linearize equation (6) and omit any dissipative terms. Combining these linearized bubble dynamics and mixture equations yields the (inviscid) equation of wave propagation in foam mixtures. Here and in the following we will write this equation in terms of a velocity potential  $\varphi$  ( $\vec{v} = \vec{\nabla}\varphi$ ) keeping in mind that all variables  $\rho'$ ,  $p'$ , and  $\varphi$  are proportional to each other in the linear approximation.

$$\frac{\partial^2 \varphi}{\partial t^2} - c_B^2 \nabla^2 \varphi - \frac{c_B^2}{\omega_B^2} \frac{\partial^2}{\partial t^2} \nabla^2 \varphi = 0, \quad (8)$$

$$c_B^2 = \frac{3\kappa p_0 + (3\kappa - 1) \frac{2\sigma}{a_0}}{3\rho_l \alpha_{g0} (1 - \alpha_{g0})},$$

$$\omega_B^2 = \frac{\omega_{SB}^2}{1 - \alpha_{g0}^{1/3}}, \quad \omega_{SB}^2 = \frac{3\kappa p_0 + (3\kappa - 1) \frac{2\sigma}{a_0}}{\rho_l a_0^2}.$$

Here  $c_B$  is the speed of sound in a wet foam.  $\omega_{SB}$  is the frequency of free oscillations of single bubble in an infinite liquid.  $\omega_B$  is the frequency of free oscillations of bubbles in a foam.

#### IV. NORMAL MODES OF A SPHERICAL FOAM DROP

Let us consider a foam sample consisting of a spherical liquid drop of radius  $R$  with  $N$  spherical gas bubbles dispersed inside. To derive formulae for the eigenfrequencies of such a foam sample, let the wave field inside the foam drop be described by

$$\varphi = \Phi \exp(i\omega t), \quad (9)$$

where  $\omega$  is the (unknown) frequency of free oscillations. Substitution of (9) in the wave equation (8) results in the Helmholtz equation for the amplitude of the velocity potential  $\Phi$

$$\nabla^2 \Phi + k^2 \Phi = 0, \quad k^2 = \frac{\omega^2}{c_B^2 \left(1 - \frac{\omega^2}{\omega_B^2}\right)}. \quad (10)$$

The oscillating surface of a foam drop can be expressed in the following way:

$$r_s = R_0 + \sum_{n=0}^{\infty} b_n P_n(\cos \theta) \exp i\omega t, \quad (11)$$

where  $P_n(\cos \theta)$  are the Legendre polynomials, and the coefficients  $b_n$  are the unknown amplitudes of eigenmodal shape oscillations ( $b_n \ll R_0$ ).

The solution of Eqs (9), (10)

$$\varphi = \sum_{n=0}^{\infty} B_n j_n(kr) P_n(\cos \theta) \exp i\omega t \quad (12)$$

describes the velocity potential inside the foam drop. Here  $j_n$  are the spherical Bessel functions, and the  $B_n$  are the unknown amplitudes for the acoustic field.

The relationship between  $b_n$  and  $B_n$  can be found by matching the normal component of the foam velocity at the drop wall with the normal displacement of the drop surface at that point, which in the linear case is

$$\left. \frac{\partial \varphi}{\partial r} \right|_{r=R_0} = \dot{r}_s. \quad (13)$$

From Eqs (12), (13) and (11) we thus obtain a formula for the velocity potential in the foam drop

$$\varphi = \sum_{n=0}^{\infty} \frac{i\omega b_n}{k} \frac{j_n(kr)}{j'_n(kR_0)} P_n(\cos \theta) \exp i\omega t. \quad (14)$$

By imposing normal stress continuity we obtain the internal pressure

$$p = p_0 + \frac{2\sigma}{R_0} - \rho_l(1 - \alpha_{g0}) \frac{\partial \varphi}{\partial t} = \quad (15)$$

$$p_0 + \frac{2\sigma}{R_0} + \rho_l(1 - \alpha_{g0}) \sum_{n=0}^{\infty} \frac{\omega^2 b_n}{k} \frac{j_n(kr)}{j'_n(kR_0)} P_n(\cos \theta) \exp i\omega t. \quad (16)$$

That pressure must be balanced with sum of the external pressure and the Laplace pressure  $p_\sigma$

$$p_\sigma = \frac{2\sigma}{R_0} + \sigma \sum_{n=0}^{\infty} \frac{(n-1)(n+2)}{R_0^2} b_n P_n(\cos \theta) \exp i\omega t. \quad (17)$$

Equating terms with the same  $n$  we find the following equation:

$$\omega_n^2 = \frac{\sigma(n-1)(n+2)k}{\rho_l(1 - \alpha_{g0})R_0^2} \frac{j'_n(kR_0)}{j_n(kR_0)}, \quad n = 0, 1, \dots \quad (18)$$

Eq. (18) together with Eq. (10) constitute a full set of equations to calculate resonance frequencies of a foam drop.

Let us consider two cases: monopole (breathing) oscillations ( $n = 0$ ) and quadrupole shape oscillations ( $n = 2$ ).

### A Breathing mode oscillations

When  $n = 0$ , Eq. (??) becomes:

$$\omega^2 = -\frac{2\sigma k}{\rho_l(1 - \alpha_{g0})R_0^2} \frac{j'_0(kR_0)}{j_0(kR_0)}. \quad (19)$$

Using an explicit expression for the spherical Bessel function of zero order ( $j_0(kR_0) = \sin(kR_0)/kR_0$ ) this equation may be presented in dimensionless form as

$$\frac{z^2}{1 + bz^2} \frac{\sin z}{z} + s \left( \cos z - \frac{\sin z}{z} \right) = 0, \quad (20)$$

$$z = kR_0, \quad b = \frac{c_B^2}{\omega_B^2 R_0^2}, \quad s = \frac{2\sigma}{\rho_l(1 - \alpha_{g0})R_0 c_B^2}.$$

For typical parameter values used in our experiments [10] the dimensionless variables  $b$  and  $s$  are very small

( $b \sim 10^{-3}$ ,  $s \sim 10^{-4}$ ). When  $b$  and  $s$  equal zero, Eq. (20) reduces to  $\sin(z) = 0$ , for which the solution is  $z = m\pi$ . That means that Eq. (20) may be used to calculate a small correction to that solution.

$$z = m\pi + \varepsilon, \quad \varepsilon \ll \pi, \quad m = 0, 1, \dots \quad (21)$$

Substitution of Eq. (21) into Eq. (20) leads to the solution

$$z = m\pi \left( 1 - \frac{1 + b\pi^2}{\pi^2} s \right), \quad (22)$$

which gives the following generalized formula for the free monopole oscillations of a foam drop (compare to Eq. (24))

$$\omega_0^2 = \frac{\omega_{SB}^2}{\frac{3}{m^2\pi^2} \left( 1 + 2 \frac{1 + b\pi^2}{\pi^2} s \right) \frac{R_0^2}{a_0^2} \alpha_{g0}(1 - \alpha_{g0}) + 1 - \alpha_{g0}^{1/3}}. \quad (23)$$

It is interesting to note that the effect of including surface tension is to lower the eigenfrequency.

For  $R_0$  large enough to ignore surface tension pressure (thus letting parameters  $b$  and  $s$  go to zero), we obtain

$$\omega^2 = \frac{\omega_{SB}^2}{\frac{3}{m^2\pi^2} \frac{R_0^2}{a_0^2} \alpha_{g0}(1 - \alpha_{g0}) + 1 - \alpha_{g0}^{1/3}}. \quad (24)$$

Eq. (24) was evaluated for the parameters of a foam drop taken from the experimental observations [10]. Namely,  $R_0 = 3.78$  mm,  $0.3$  mm  $< a_0 < 0.5$  mm. The normalized frequencies  $\omega/\omega_{SB}$  versus foam void fraction  $\alpha_{g0}$  for  $m = 1$  are shown in Fig. 1. The dashed line denotes a gas volume fraction  $\alpha_{g0} = 0.77$  of the foam drop used in [10]. One can see that the monopole frequency of the foam drop asymptotes to the single bubble result  $\omega_{SB}$  in the low void fraction limit, and to infinity for a void fraction of unity. In between these limiting values, the foam drop frequency is some fraction of  $\omega_{SB}$ .

Typically,  $R_0 \gg a_0$ , and Eq. (24) may be simplified as follows

$$\omega^2 \approx \frac{\omega_{SB}^2}{\frac{3}{m^2\pi^2} \frac{R_0^2}{a_0^2} \alpha_{g0}(1 - \alpha_{g0})}. \quad (25)$$

Thus, with the Eq. (8) and taking into account that  $2\sigma/a_0 \ll p_0$  one can get the formula for the frequency of the foam drop monopole oscillations, which looks similar to the well-known Minnaert formula [31] for single bubble monopole oscillations

$$\omega^2 \approx \frac{3\kappa p_0}{\rho_l a_*^2}, \quad a_* = \frac{\sqrt{3\alpha_{g0}(1 - \alpha_{g0})}}{m\pi} R_0. \quad (26)$$

Here  $a_*$  is an effective bubble radius. It is easy to estimate that for the experimental data ( $R_0 = 3.78$  mm,  $\alpha_{g0} = 0.77$ ,  $0.3$  mm  $< a_0 < 0.5$  mm) the effective bubble radius  $a_* = 0.88$  mm is larger than bubble radius and smaller than radius of the foam drop ( $a_0 < a_* < R_0$ ).

These results, especially Eq. (24) may be usefully compared to previous results for the breathing mode of a compact bubble cloud in water derived by several authors [32] [35]. In those works, the motivation was to explain low frequency ambient noise in the ocean as due to collective oscillations of clouds of bubbles. The low void fraction limit is the same, but the high void fraction limit is not, since in the present case the high void fraction limit corresponds to the effective mass of the oscillator approaching zero, whereas in the bubble cloud scenario, the high void fraction limit corresponds to a single bubble of radius  $R$ .

It is remarkable that (approximately) the frequency does not depend on bubble radius, but depends only on the void fraction of the foam and the radius of the foam drop. It is clear that the apparent difference between frequencies for different bubble radii shown in Fig. 1 is only due to differences in the single bubble frequency  $\omega_{SB}$  by which we normalize. For instance, when  $\alpha_{g0} = 0.77$  (dash line) the frequencies of monopole oscillations for  $a_0 = 0.3$  mm and  $a_0 = 0.5$  mm are equal to 3708 Hz and 3674 Hz respectively. This strengthens the case for our working assumption that the most important parameter is the void fraction - the details of the bubble size distribution should not affect the leading order results.

## B Shape mode oscillations

When  $n = 2$  Eq. (??) becomes:

$$\omega_2^2 = \frac{4\sigma k}{\rho_l(1 - \alpha_{g0})R_0^2} \frac{j_2'(kR_0)}{j_2(kR_0)}. \quad (27)$$

This equation may be presented in dimensionless form as

$$\frac{z}{1 + bz^2} = 2s \frac{j_2'(z)}{j_2(z)}, \quad (28)$$

where dimensionless variables  $z, b, s$  are defined in Eq. (20).

To look for the low-frequency solution of this equation one should use the asymptotic expansion for the spherical Bessel function of second order when  $z \rightarrow 0$  [36],

$$j_2(z) \approx \frac{z^2}{15} \left(1 - \frac{z^2}{14}\right), \quad j_2'(z) \approx \frac{2}{15}z - \frac{2}{105}z^3. \quad (29)$$

Then Eq. (28) may be rewritten in the following form

$$\frac{z}{1 + bz^2} = \frac{4s}{z} \left(1 - \frac{z^2}{14}\right). \quad (30)$$

There is one low-frequency solution of this equation which is approximately equal to

$$z^2 = 4s \left[1 - 4s \left(\frac{1}{14} - b\right)\right]. \quad (31)$$

In terms of  $\omega$ ,

$$\omega_2^2 = \omega_{L2}^2 \left\{1 - \frac{12\sigma\alpha_{g0}}{7R_0 \left[3\kappa p_0 + (3\kappa - 1)\frac{2\sigma}{a_0}\right]}\right\}. \quad (32)$$

$$\omega_{L2}^2 = \frac{8\sigma}{\rho_l(1 - \alpha_{g0})R_0^3}. \quad (33)$$

Here  $\omega_{L2}$  is the Lamb frequency of quadrupole oscillations for an inviscid and incompressible liquid drop [37] of density  $\rho_l(1 - \alpha_{g0})$  surrounded by a vacuum. In our case  $\omega_{L2} \approx 210$  s<sup>-1</sup>. Eq. (32) presents a correction to that frequency due to foam compressibility. For our experimental parameters the correction term is negligibly small ( $\sim 10^{-4}$ ).

## V. COUPLING OF BREATHING AND SHAPE OSCILLATIONS

Here we consider the influence of breathing ('monopole' or 'radial') oscillations on the quadrupole mode. This corresponds to an experimental situation in which the breathing mode is directly excited by an external field with pressure gradients large compared to the drop radius  $R$ .

It is instructive to compare the period of quadrupole oscillations with the time required for an acoustic wave to cross the foam drop. At frequencies about 200 s<sup>-1</sup> (see  $\omega_{L2}$  in the previous section) the period of oscillations is about  $3 \cdot 10^{-2}$  s. The speed of sound for our foam is  $c_B \sim 30$  m/s. Then for a foam drop with radius  $R = 3.78$  mm, the acoustic transit time is about  $10^{-4}$  s which is much less than the period of the quadrupole mode. Thus, one can assume the foam drop liquid pressure  $p_d$  to be uniform in space and variable in time, as was done for individual bubbles in section 1 above.

As in Section 3, each single bubble in the bubbly liquid is placed in the center of spherical cell with the radius  $A$  chosen in such a way as to cover the whole bubbly mixture by cells. The only difference is that in Section 3 all variables were functions of time and space coordinates. In the present case, all these variables depend only on time. Thus, Eqs (2), (3), (5), (7) are valid, but for temporal variations only.

Taking into account that the pressure of the two-phase mixture  $p_d$  is uniform in space and time-dependent, one

can get the following generalized Rayleigh-Plesset equation for radial bubble motion in a cell (compare to Eq. (6)):

$$\left(1 - \alpha_g^{1/3}\right) a\ddot{a} + \frac{3}{2} \left(1 - \frac{4}{3}\alpha_g^{1/3} + \frac{1}{3}\alpha_g^{4/3}\right) \dot{a}^2 = \frac{p_a - p_d}{\rho_l}. \quad (34)$$

Conservation of the (incompressible) liquid volume in the foam drop leads to the following simple relationship between bubble and drop radii.

$$R^3 - R_0^3 = N(a^3 - a_0^3). \quad (35)$$

Eqs. (3),(5)-(7),(refR-P-1), and (35) form a complete nonlinear system that describes radial motion due to foam drop oscillations.

In case of weak (linear) oscillations the system may be reduced to a forced linear oscillator:

$$\ddot{\tilde{R}} + \omega_B^2 \tilde{R} = -F \sin \omega t, \quad (36)$$

$$F = \frac{\alpha_{g0} \Delta p}{\rho_l \left(1 - \alpha_{g0}^{1/3}\right) a_0^2}.$$

Here  $\tilde{R} = (R - R_0)/R_0$  is the relative foam drop radius fluctuation,  $\omega_B$  is the frequency of free oscillations of bubbles in foam (see Eq. (8)),  $\Delta p$  is an external driving ( $p_d = p_0 + \Delta p \sin \omega t$ ). The Laplace pressure on the drop wall is neglected.

In order to investigate coupling between breathing and shape oscillations it is useful to consider the radial breathing motion as the base motion (below marked by an asterisk) and shape oscillations as a perturbation of this base motion (below marked by a tilde). In the homobaric approximation discussed above, the velocity distribution  $w_*(r, t)$  for monopole oscillations (which satisfies the mass balance equation for the foam mixture) is given by

$$w_* = \frac{\dot{R}}{R} r. \quad (37)$$

The dynamic drop shape is expressed by the expansion

$$r_s = R(t) + \sum_{n=1}^{\infty} b_n(t) P_n(\cos \theta), \quad (38)$$

where  $P_n(\cos \theta)$  are the Legendre polynomials,  $R$  is the instantaneous foam drop radius and the amplitudes of eigenmodal shape oscillations  $b_n$  ( $b_n \ll R_0$ ) are unknown functions of time. Our goal is to derive a coupled set of differential equations for the variables  $R(t), b_n(t)$ .

The velocity distribution and potential inside the foam drop are:

$$w = \frac{\partial \varphi}{\partial r} = w_* + \tilde{w} = \frac{\dot{R}}{R} r + \sum_{n=1}^{\infty} n B_n(t) r^{n-1} P_n(\cos \theta) \quad (39)$$

$$\varphi = \varphi_* + \tilde{\varphi} = \frac{\dot{R} r^2}{2R} + \sum_{n=1}^{\infty} B_n(t) r^n P_n(\cos \theta) \quad (40)$$

Here  $B_n(t)$  are the unknown amplitudes of the foam mixture motion due to shape oscillations.

The relationship between  $b_n(t)$  and  $B_n(t)$  can be found by matching the normal component of the foam velocity at the drop wall with the normal displacement of the drop surface at that point, which in the linear approximation is

$$w|_{r=r_s} = \dot{r}_s. \quad (41)$$

From Eqs (38),(39) and (41) we thus obtain the formulae for the velocity and velocity potential in the foam drop

$$\varphi = \frac{\dot{R} r^2}{2R} + \tilde{\varphi}, \quad \tilde{\varphi} = \sum_{n=1}^{\infty} \left( \dot{b}_n - \frac{\dot{R}}{R} b_n \right) \frac{r^n}{n R^{n-1}} P_n(\cos \theta), \quad (42)$$

$$w = \frac{\dot{R}}{R} r + \tilde{w}, \quad \tilde{w} = \sum_{n=1}^{\infty} \left( \dot{b}_n - \frac{\dot{R}}{R} b_n \right) \frac{r^{n-1}}{R^{n-1}} P_n(\cos \theta). \quad (43)$$

In order to derive an approximate equation for drop shape oscillations let us consider the momentum equation for the foam mixture,

$$\rho \frac{d\vec{v}}{dt} + \vec{\nabla} p = 0, \quad (44)$$

where  $d/dt$  is the material derivative, and  $\vec{v}$  is the velocity of the mixture (liquid and bubbles move with equal velocities). We now take a linearized form of the radial component of (44) as follows.

$$\rho_* \left( \frac{\partial \tilde{w}}{\partial t} + w_* \frac{\partial \tilde{w}}{\partial r} + \tilde{w} \frac{\partial w_*}{\partial r} \right) + \frac{\partial \tilde{p}}{\partial r} = 0. \quad (45)$$

Here  $\rho_* = \rho_l(1 - \alpha_g)$  is a function of time. Integration over the radial space coordinate  $r$  leads to the following specific form of Bernoulli's integral

$$\rho_* \left( \frac{\partial \tilde{\varphi}}{\partial t} + w_* \frac{\partial \tilde{\varphi}}{\partial r} \right) + \tilde{p} = \Psi(t). \quad (46)$$

The function  $\Psi(t)$  may be determined by evaluating the left-hand-side of Eq. (46) at the limits  $r = r_s$  and  $r = 0$ . We obtain approximately

$$\left. \frac{\partial \tilde{\varphi}}{\partial t} \right|_{r=r_s} \approx \sum_{n=1}^{\infty} \frac{R^n}{n} \frac{d}{dt} \left[ \frac{1}{R^{n-1}} \left( \dot{b}_n - \frac{\dot{R}}{R} b_n \right) \right] P_n(\cos \theta),$$

$$\left. \frac{\partial \tilde{\varphi}}{\partial r} \right|_{r=r_s} \approx \sum_{n=1}^{\infty} \left( \dot{b}_n - \frac{\dot{R}}{R} b_n \right) P_n(\cos \theta), \quad w_s|_{r=r_s} \approx \dot{R},$$

$$\tilde{p}|_{r=r_s} \approx \sigma \sum_{n=1}^{\infty} \frac{(n-1)(n+2)}{R^2} b_n P_n(\cos \theta).$$

Then Eq. (??) leads to the following equation for the evolution of the amplitudes of the axisymmetric modes

$$\ddot{b}_n + \left[ \frac{(n-1)n(n+2)\sigma}{\rho_* R^3} - \frac{\ddot{R}}{R} \right] b_n = 0, \quad (47)$$

which taking into account that  $\rho_* R^3 = \rho_l(1 - \alpha_{g0})R_0^3$  may be rewritten as follows

$$\ddot{b}_n + \left( \omega_{Ln}^2 - \frac{\ddot{R}}{R} \right) b_n = 0, \quad \omega_{Ln}^2 = \frac{(n-1)n(n+2)\sigma}{\rho_l(1 - \alpha_{g0})R_0^3}, \quad (48)$$

where  $\omega_{Ln}$  is the Lamb frequency of shape oscillations of a foam drop with mode number  $n$  and effective density  $\rho_l(1 - \alpha_{g0})$ . This result is analogous to the well-studied problem of parametric shape oscillations of single bubbles in an infinite fluid, see for example [38] and [39].

To obtain an analytical expression, if we consider only linear foam drop monopole oscillations, then the solution of Eq. (36) is

$$\tilde{R} = -\frac{F \sin \omega t}{\omega_B^2 - \omega^2}, \quad (49)$$

and Eq. (48) may be presented in the form of Mathieu's equation.

$$\frac{d^2 b_n}{d\tau^2} + (K - 2\varepsilon \sin 2\tau) b_n = 0, \quad (50)$$

$$K = \frac{4\omega_{Ln}^2}{\omega^2}, \quad \varepsilon = \frac{2F}{\omega_B^2 - \omega^2}, \quad \tau = \frac{\omega t}{2}.$$

Solutions of Eq. (50) are well studied (see [36]). There exist two types of solutions in the inviscid limit: bounded (stable) and unbounded (unstable) in time.

## VI. NUMERICAL RESULTS

### A Nonlinear breathing dynamics

TBD

### B Parametric instability

It is beyond the scope of this work to investigate dissipative effects. Nevertheless in the low void fraction limit we may incorporate the effects of weak liquid viscosity into the equation of motion for the shape oscillations described by Eq. (48). We give here without derivation the parametric equation of motion including weak damping of the shape oscillations:

$$\ddot{b}_n + \left( \frac{3\dot{R}}{R} + 2\beta_{Ln} \right) \dot{b}_n + \left( \omega_{Ln}^2 - \frac{\ddot{R}}{R} \right) b_n = 0, \quad \beta_{Ln} = \frac{(n-1)(2n+1)}{\rho_l(1 - \alpha_{g0})R_0^3} \quad (51)$$

where  $\beta_{Ln}$  is the classical expression for the weak viscous damping of the shape oscillations of a pure liquid drop. We integrate Eq. (51) simultaneously with Eqs (34),(5)-(7),(35) to investigate the effect of a time-varying void fraction on the stability of the spherical foam drop.

The regions of parameter space  $(K, \varepsilon)$  where one obtains stable or unstable parametric shape oscillations are shown on Fig.2, where stable zones are colored in black. One can see that the optimal driving frequency leading to parametric instability is two times larger than the eigenfrequency of the shape oscillations ( $K = 1$ ), the 2:1 external resonance.

In Fig. 3 we plot the frequency of quadrupole oscillations  $\omega_{L2}/2\pi$  and the parametric driving resonance frequency  $\omega = \omega_r = 2\omega_{L2}$  versus foam void fraction  $\alpha_{g0}$ . For our experiments, the  $\omega_{(2)} \sim 210 \text{ s}^{-1}$ , the frequency of parametric resonance is  $\omega_r \approx 420 \text{ s}^{-1}$ . It corresponds to a frequency  $f = \omega/2\pi \approx 65 \text{ Hz}$ , which was observed in experiments [10].

## VII. DISCUSSION

A new bubble-based mathematical model of foam drop dynamics is presented. According to this model different types of foam drop oscillations are analyzed. The derived formulae are used to explain the experimental data published recently [10]. In that work, an experimental technique was demonstrated for acoustically levitating aqueous foam drops and acoustically exciting their spheroidal modes. Results were presented in which a foam drop with gas volume fraction  $\alpha_{g0} = 0.77$  was levitated at 30 KHz and excited into a quadrupole resonance at  $63 \pm 3 \text{ Hz}$ .

Here it is shown that monopole (radial) oscillations and quadrupole (shape) oscillations, for foam parameters taken from experimental observations, are characterized



by two different scales of frequencies: about 3700 Hz for monopole oscillations, and about 33 Hz for quadrupole oscillations.

Monopole frequency, being far from both, levitation frequency and quadrupole frequency, probably does not play any role in the phenomenon. The quadrupole oscillation may be excited by parametric resonance mechanism. The highest frequency of the parametric excitation is about 65 Hz, which corresponds with experimental data [10] very well.

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